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Reflection from a Bianisotropic Dielectric

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Abstract

The well-known surface densities of bound charge $\sigma = \mathbf{P} \cdot \mathbf{n}$ and current $\mathbf{K} = \mathbf{M} \times \mathbf{n}$, which give rise to discontinuities in fields at a surface, are inconsistent in their multipole order. The electric quadrupole contribution is missing from each. A consequence of including this is the appearance of a surface density of electric dipole moment $\boldsymbol{\mathcal{P}}$. Its effect on the boundary conditions is derived, and a reflection experiment on a bianisotropic crystal is proposed, which in conjunction with a transmission experiment, allows the $\boldsymbol{\mathcal{P}}$ contribution to be measured.

1. Introduction

The contribution to a physical effect of electric quadrupoles induced in matter by long wavelength radiation is known to compare in magnitude with that of induced magnetic dipoles [1, 2, 3]. Consequently, there is an inconsistency in the surface densities of bound charge and current [4]

$$\sigma = \mathbf{P} \cdot \mathbf{n}, \qquad \mathbf{K} = \mathbf{M} \times \mathbf{n}, \tag{1}$$

where $\mathbf{n}$ is the outward unit normal at a macroscopic point on the surface and $\mathbf{P}$ and $\mathbf{M}$ are the electric and magnetic dipole moments per unit macroscopic volume. An electric quadrupole term should be included in $\mathbf{K}$ alongside $\mathbf{M}$, and also one in σ.

This paper derives these two contributions and then shows their effect on the boundary conditions on the fields at a vacuum-dielectric interface. In particular, the electric dipole surface density also emerges and its role is examined, as well as its contribution to a physical effect.

2. Theory of Bound Source Contributions

The retarded vector potential outside a bounded distribution of currents in vacuum is [4]

$$\mathbf{A}(\boldsymbol{\mathcal{R}}, t) = (\mu_0/4\pi) \int_v \mathbf{J}(\mathbf{r}, t - |(\boldsymbol{\mathcal{R}} - \mathbf{r}|/c)/|(\boldsymbol{\mathcal{R}} - \mathbf{r}|dv. \tag{2}$$

By expanding $\mathbf{J}$ for $\mathcal{R} \gg r$, applying the condition $\lambda \gg \mathcal{R}$ and averaging over a macroscopic volume element dV, we obtain the macroscopic vector potential due to a bounded dielectric of volume V and surface area S. To order electric quadrupole (E2)-magnetic dipole (M1) it is [5]

$$A_\alpha(\mathbf{R}, t) = (\mu_0/4\pi)\{\int_V (1/R)(\dot{P}_\alpha - \tfrac{1}{2}\nabla_\beta \dot{Q}_{\alpha\beta} + \epsilon_{\alpha\beta\gamma}\nabla_\beta M_\gamma)dV + \int_S (1/R)(\tfrac{1}{2}\dot{Q}_{\alpha\beta} - \epsilon_{\alpha\beta\gamma}M_\gamma)n_\beta da\}. \tag{3}$$

In this $\mathbf{R} = -\boldsymbol{\mathcal{R}}$ is the displacement of dV from the field point and $Q_{\alpha\beta} = \sum q r_\alpha r_\beta / dV$ is the electric quadrupole moment per unit volume. The multipole moment densities P_α, M_α, $Q_{\alpha\beta}$ are relative to an arbitrary origin in dV at the time $t - R/c$. Similarly for the scalar potential [5]

$$
\begin{aligned}
\phi(\mathbf{R},t) &= 1/(4\pi\epsilon_0)\left\{\int_V (1/R)\nabla_\alpha(-P_\alpha + \tfrac{1}{2}\nabla_\beta Q_{\alpha\beta})dV\right. \\
&+ \left.\int_S [(1/R)\{(P_\alpha - \tfrac{1}{2}\nabla_\beta Q_{\alpha\beta})n_\alpha + \nabla_{||}\mathcal{P}_{||}\} + (R_\perp/R^3)\mathcal{P}_\perp]da\right\}, \qquad (4)
\end{aligned}
$$

where $\nabla_{||}$ denotes differentiation parallel to the surface element da and

$$\mathcal{P}_\alpha = -\tfrac{1}{2}Q_{\alpha\beta}n_\beta \qquad (5)$$

is a contribution of E2 order. From the volume integrals in (3) and (4), with their $1/R$ dependence, the bound source volume densities of current and charge are

$$J_\alpha = \dot{P}_\alpha - \tfrac{1}{2}\nabla_\beta\dot{Q}_{\alpha\beta} + \epsilon_{\alpha\beta\gamma}\nabla_\beta M_\gamma, \quad \rho = -\nabla_\alpha P_\alpha + \tfrac{1}{2}\nabla_\alpha\nabla_\beta Q_{\alpha\beta}. \qquad (6)$$

These satisfy the equation of continuity for bound sources $\nabla\cdot\mathbf{J} = -\dot{\rho}$. From the $1/R$ terms in the surface integrals in (3) and (4) the surface densities of bound current and charge are

$$K_\alpha = (\tfrac{1}{2}\dot{Q}_{\alpha\beta} - \epsilon_{\alpha\beta\gamma}M_\gamma)n_\beta, \quad \sigma = (P_\alpha - \tfrac{1}{2}\nabla_\beta Q_{\alpha\beta})n_\alpha + \nabla_{||}\mathcal{P}_{||}. \qquad (7)$$

There still remains in the surface integral in (4) the term with a $\mathbf{R}/R^3$ dependence. As this is characteristic of the potential due to an electric dipole, we interpret $\boldsymbol{\mathcal{P}}$ in (4) as the surface density of electric dipole moment due to bound charge on the surface.

If the bound sources in (6) are used in the two inhomogeneous Maxwell equations

$$\nabla\cdot\mathbf{E} = (\rho_c + \rho)/\epsilon_0, \quad \nabla\times\mathbf{B} = \mu_0(\epsilon_0\dot{\mathbf{E}} + \mathbf{J}_c + \mathbf{J}), \qquad (8)$$

where ρ_c and $\mathbf{J}_c$ are the corresponding free source terms, one obtains by comparison with

$$\nabla\cdot\mathbf{D} = \rho_c, \quad \nabla\times\mathbf{H} = \dot{\mathbf{D}} + \mathbf{J}_c, \qquad (9)$$

the multipole expressions for $\mathbf{D}$ and $\mathbf{H}$ to order E2–M1

$$D_\alpha = \epsilon_0 E_\alpha + P_\alpha - \tfrac{1}{2}\nabla_\beta Q_{\alpha\beta}, \quad H_\alpha = B_\alpha/\mu_0 - M_\alpha. \qquad (10)$$

The densities P_α, M_α, $Q_{\alpha\beta}$ are induced by the wave fields and their space and time derivatives

$$E_\alpha,\ \dot{E}_\alpha,\ \nabla_\beta E_\alpha,\ \nabla_\beta\dot{E}_\alpha,\ \ldots;\quad B_\alpha,\ \dot{B}_\alpha,\ \nabla_\beta B_\alpha,\ \nabla_\beta\dot{B}_\alpha,\ \ldots. \qquad (11)$$

Then to E2–M1 order

$$
\begin{aligned}
P_\alpha &= \alpha_{\alpha\beta}E_\beta + \omega^{-1}\alpha'_{\alpha\beta}\dot{E}_\beta + \tfrac{1}{2}a_{\alpha\beta\gamma}\nabla_\gamma E_\beta + \tfrac{1}{2}\omega^{-1}a'_{\alpha\beta\gamma}\nabla_\gamma\dot{E}_\beta + G_{\alpha\beta}B_\beta + \omega^{-1}G'_{\alpha\beta}\dot{B}_\beta, \qquad (12) \\
Q_{\alpha\beta} &= a_{\gamma\alpha\beta}E_\gamma + \omega^{-1}a'_{\gamma\alpha\beta}\dot{E}_\beta, \qquad (13) \\
M_\alpha &= G_{\beta\alpha}E_\beta - \omega^{-1}G'_{\beta\alpha}\dot{E}_\beta, \qquad (14)
\end{aligned}
$$

where ω is the angular frequency. From their definitions $\mathbf{E}$, $\boldsymbol{\nabla}$, $\mathbf{P}$, $Q_{\alpha\beta}$ are time-even and $\mathbf{B}$, $\mathbf{M}$ time-odd. Thus $\alpha'_{\alpha\beta}$, $a'_{\alpha\beta\gamma}$, $G_{\alpha\beta}$ are time-odd and belong only to magnetic crystals. With (13)–(14) in (10), constitutive relations for $\mathbf{D}$ and $\mathbf{H}$ are obtained. However, these do not satisfy Post's covariance requirement for a plane time-harmonic wave for negligible absorption [6]

$$D_\alpha = A_{\alpha\beta}E_\beta + T_{\alpha\beta}B_\beta, \quad H_\alpha = U_{\alpha\beta}E_\beta + X_{\alpha\beta}B_\beta, \quad A_{\alpha\beta} = A^*_{\beta\alpha}, \quad X_{\alpha\beta} = X^*_{\beta\alpha}, \quad U_{\alpha\beta} = -T^*_{\beta\alpha}. \qquad (15)$$

From $\nabla \times \mathbf{E} = -\dot{\mathbf{B}}$ and with (10), (12)–(14) substituted into (9), one can show that

$$\begin{aligned} P_\alpha &= \alpha_{\alpha\beta}E_\beta + \omega^{-1}[\alpha'_{\alpha\beta}]\dot{E}_\beta + \tfrac{1}{6}\omega^{-1}[a'_{\alpha\beta\gamma} + a'_{\beta\gamma\alpha} + a'_{\gamma\alpha\beta}]\nabla_\gamma\dot{E}_\beta \\ &+ [G_{\alpha\beta} - \tfrac{1}{3}\delta_{\alpha\beta}G_{\gamma\gamma} - \tfrac{1}{6}\omega\epsilon_{\beta\gamma\delta}a'_{\gamma\delta\alpha}]B_\beta + \omega^{-1}(G'_{\alpha\beta} - \tfrac{1}{2}\omega\epsilon_{\beta\gamma\delta}a_{\gamma\delta\alpha})\dot{B}_\beta, && (16) \\ Q_{\alpha\beta} &= -\tfrac{1}{3}\omega^{-1}[a'_{\alpha\beta\gamma} + a'_{\beta\gamma\alpha} + a'_{\gamma\alpha\beta}]\dot{E}_\gamma = -S_{\alpha\beta\gamma}\dot{E}_\gamma, && (17) \\ M_\alpha &= [G_{\beta\alpha} - \tfrac{1}{3}\delta_{\alpha\beta}G_{\gamma\gamma} - \tfrac{1}{6}\omega\epsilon_{\alpha\gamma\delta}a'_{\gamma\delta\beta}]E_\beta - \omega^{-1}(G'_{\beta\alpha} - \tfrac{1}{2}\omega\epsilon_{\alpha\gamma\delta}a_{\gamma\delta\beta})\dot{E}_\beta. && (18) \end{aligned}$$

Terms in brackets [ ] are time-odd. From (10), (16)–(18) and with $\mathbf{E} = \mathbf{E}_0 \exp\{-i(\omega t - \mathbf{k}\cdot\mathbf{r})\}$ the covariant forms in (15) are satisfied. Then from (6) the Ampère-Maxwell equation in (8) is

$$\epsilon_{\alpha\beta\gamma}\nabla_\beta B_\gamma + i\omega\mu_0\epsilon_{\alpha\beta}E_\beta = 0 \tag{19}$$

for a dielectric, where $\epsilon_{\alpha\beta}$ is the dynamic permittivity tensor, which to order E2–M1 is

$$\begin{aligned} \epsilon_{\alpha\beta} &= \epsilon_0\delta_{\alpha\beta} + \alpha_{\alpha\beta} - i\alpha'_{\alpha\beta} + \omega^{-1}k_\gamma(A_{\alpha\beta\gamma} - iA'_{\alpha\beta\gamma}), && (20) \\ A_{\alpha\beta\gamma} &= -\epsilon_{\beta\gamma\delta}G_{\alpha\delta} - \epsilon_{\alpha\gamma\delta}G_{\beta\delta} + \tfrac{1}{2}\omega(a'_{\alpha\beta\gamma} + a'_{\beta\alpha\gamma}), && (21) \\ A'_{\alpha\beta\gamma} &= -\epsilon_{\beta\gamma\delta}G'_{\alpha\delta} + \epsilon_{\alpha\gamma\delta}G'_{\beta\delta} - \tfrac{1}{2}\omega(a_{\alpha\beta\gamma} - a_{\beta\alpha\gamma}). && (22) \end{aligned}$$

We now show that the surface discontinuities in (7) alter the usual Maxwell boundary conditions.

3. The Boundary Conditions

To derive these, unit step functions are used instead of the integral forms of Maxwell's equation. With the surface element in the xy plane and $+z$ axis into the medium, these functions and their derivatives are, using the Dirac δ-function and its derivative δ',

$$u(z) = \begin{cases} 1 \text{ for } z > 0 \\ 0 \text{ for } z < 0 \end{cases}, \quad u(-z) = \begin{cases} 0 \text{ for } z > 0 \\ 1 \text{ for } z < 0 \end{cases}, \quad \frac{\partial u(\pm z)}{\partial z} = \pm\delta(z), \quad \frac{\partial^2 u(\pm z)}{\partial z^2} = \pm\delta'(z), \tag{23}$$

The total bound current and charge densities are then

$$\begin{aligned} \mathbf{J}_t(\mathbf{R}) &= u(z)\mathbf{J}_1(\mathbf{R}) + u(-z)\mathbf{J}_2(\mathbf{R}) + \delta(z)\mathbf{K}(\mathbf{r}), && (24) \\ \rho_t(\mathbf{R}) &= u(z)\rho_1(\mathbf{R}) + u(-z)\rho_2(\mathbf{R}) + \delta(z)\sigma(\mathbf{r}) + \delta'(z)\hat{\mathbf{z}}\cdot\boldsymbol{\mathcal{P}}(\mathbf{r}), && (25) \end{aligned}$$

where medium 2 is the vacuum, $\mathbf{n} = -\hat{\mathbf{z}}$, and $\mathbf{r}$ lies in the xy plane. Using (23)–(25) in $\nabla\cdot\mathbf{J} = -\dot{\rho}$ and equating the coefficients of $\delta(z)$ one obtains σ in (7) and of $\delta'(z)$ one finds

$$K_z = -\dot{\mathcal{P}}_z = \tfrac{1}{2}\dot{Q}_{zz}, \tag{26}$$

which is $\mathbf{K}$ in (7) for $\alpha = z$. Since these two results confirm (24) and (25), we similarly take

$$\begin{aligned} \mathbf{E}(\mathbf{R}) &= u(z)\mathbf{E}_1(\mathbf{R}) + u(-z)\mathbf{E}_2(\mathbf{R}) + \delta(z)\boldsymbol{\mathcal{E}}(\mathbf{r}), && (27) \\ \mathbf{B}(\mathbf{R}) &= u(z)\mathbf{B}_1(\mathbf{R}) + u(-z)\mathbf{B}_2(\mathbf{R}) + \delta(z)\boldsymbol{\mathcal{B}}(\mathbf{r}), && (28) \end{aligned}$$

where $\boldsymbol{\mathcal{E}}(\mathbf{r})$ and $\boldsymbol{\mathcal{B}}(\mathbf{r})$ are surface fields. With these in (8) one obtains the boundary conditions

$$\begin{aligned} E_{1x} - E_{2x} &= \nabla_x\mathcal{E}_z, \quad E_{1y} - E_{2y} = \nabla_y\mathcal{E}_z, \quad E_{1z} - E_{2z} = (\sigma - \epsilon_{1\alpha z}\nabla_{\Sigma\alpha}\mathcal{E}_z)/\epsilon_0, && (29) \\ B_{1x} - B_{2x} &= \mu_0(K_y + \epsilon_{1yz}\dot{\mathcal{E}}_z), \quad (B_{1y} - B_{2y}) = -\mu_0(K_x + \epsilon_{1xz}\dot{\mathcal{E}}_z), \quad B_{1z} = B_{2z}, && (30) \end{aligned}$$

$$\mathcal{E}_z = \mathcal{P}_z/\epsilon_{1zz} = -\tfrac{1}{2}Q_{zz}/\epsilon_{1zz}. \tag{31}$$

Thus to order E2–M1 the tangential components of $\mathbf{E}$ are no longer continuous across a boundary, whereas the normal component of $\mathbf{B}$ is.

4. The Role of the Surface Density of Electric Dipole Moment

As is evident from (29)–(31) the boundary conditions reduce to the Maxwell forms when $\mathcal{P}$ (or $Q_{\alpha\beta}$) is zero. In the electric dipole (E1) order $Q_{\alpha\beta} = 0$. Thus the Maxwell conditions are an approximation, applying only in the E1 order. From (5) and (17), $\mathcal{P}$ exists only for magnetic media. Also, the tensor $S_{\alpha\beta\gamma}$ in (17) may vanish for symmetry reasons, as for propagation along the main axis of antiferromagnetic Cr_2O_3, even though it possesses the time-odd tensor $a'_{\alpha\beta\gamma}$.

5. Application

The simplest magnetic crystal is an antiferromagnet (since $\alpha'_{\alpha\beta} = 0$) that is centrosymmetric (since its time-even odd-rank polar tensor $a_{\alpha\beta\gamma}$ and even-rank axial tensor $G'_{\alpha\beta}$ vanish). The only such symmetry for which an effect exists at normal incidence is $\underline{4}/\underline{m}m\underline{m}$. Its Fresnel reflection matrix for normal incidence parallel to its C_2 axis can be shown to be [7]

$$R = \begin{bmatrix} (n_o - 1)/(n_o + 1) & r_{ps} \\ r_{sp} & -(n_e - 1)/(n_e + 1) \end{bmatrix}, \tag{32}$$

where n_o and n_e are the ordinary and extraordinary refractive indices,

$$r_{sp} = r_{ps} = \mu_0 c[Kn_e/(n_o + n_e) - S_{123}]/(n_o + 1)(n_e + 1), \quad K = 2G_{11} + \omega(a'_{123} + a'_{312}), \tag{33}$$

and S_{123} is the only component that exists of the tensor in (17) which, because of (5), is the surface electric dipole term. The components in (33) are relative to crystallographic axes. The matrix in (32) is identical in form to that of Cr_2O_3 for the same geometry. The effect in Cr_2O_3 is a rotation of the plane of linearly polarized light, which has previously been measured, yielding an experimental value of r_{sp}. Thus r_{sp} should be measurable for a $\underline{4}/\underline{m}m\underline{m}$ crystal. A different combination of K and S_{123} enters the birefringence in transmission

$$n_+ - n_- = c\mu_0(3S_{123} - K). \tag{34}$$

From these reflection and transmission experiments a value can be obtained of S_{123} and hence of the contribution of the surface density of electric dipole moment $\mathcal{P}$ to the boundary conditions. This would provide a test of our theory.

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